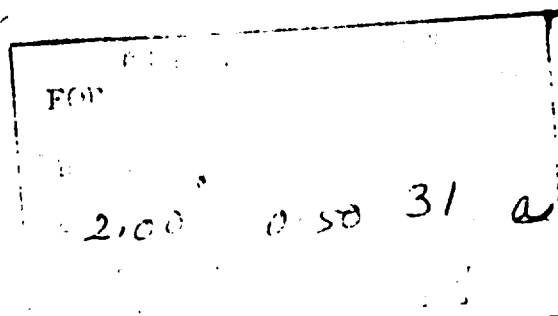USNRDL-TR-913

30 September 1965

A LABORATORY APPARATUS FOR MEASURING THE AMOUNT OF TRITIUM ACTIVITY REMOVED FROM A CONTAMINATED SURFACE, BY DIRECT CONTACT WITH THE SURFACE

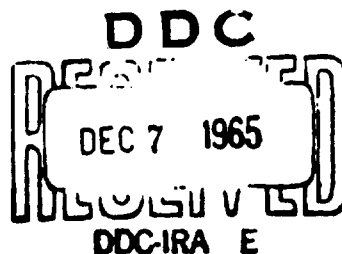by

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ABSTRACT

An apparatus is described that can be used to obtain data for evaluation of the health hazard to personnel who directly contact a surface contaminated with tritium. The sampling component is a small glass roller wrapped with a strip of filter paper dampened with ethylene glycol. It is rolled over the contaminated surface to sample the activity. The sampling apparatus has adjustments to vary the surface contact time, the area of surface sampled, and the pressure applied. After a run, the contaminated roller is dropped immediately after contact with the surface into a counting vial containing a scintillant solution, and the activity is assayed. The apparatus is small enough to be operated in a conventional glove box.

The precision of the measurements is influenced by the texture of the surface sampled. Precision measured on a smooth optical quartz surface using a nonvolatile tracer, Pm^{143}, as a contaminant was determined to be $\pm$ 2 %. The precision for tritium sampling from a relatively coarser, nonuniform metal surface was approximately $\pm$ 25 %.

The apparatus can be used to study the rate of tritium desorbed from the surface, which allows estimation of the cumulative hazard effect of multiple contacts with the surface during the desorption period. Representative samples of metal surfaces exposed to tritium show that after exposure, desorption is exponential, decreasing to an "equilibrium" amount within a few hours. Multiple contacts with these desorbing surfaces did not seem to significantly alter the desorption process.

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SUMMARY

The Problem

To design, construct and evaluate an apparatus that can be used to measure the factors required to reproducibly determine the amount of tritium activity removed from a contaminated surface by direct contact with the surface.

The Apparatus

An apparatus has been constructed to uniformly and reproducibly sample a flat firm surface. Factors such as area sampled, contact time and contact pressure can be regulated and varied. Surface-contact desorption phenomena can be determined by repeated sampling of a given area of surface as a function of time.

Findings

The precision of replicate measurements was 2 % for an extremely smooth surface such as optical quality quartz using a non-volatile activity, Pm^{143}. For relatively coarser nonuniform surfaces such as metals the precision for tritium removal is $\pm$ 25 %. The tritium activities were assayed by liquid scintillation counting techniques.

Tritium was found to desorb exponentially from various unpainted and painted surfaces. A desorption equilibrium was attained within a few hours for all the surfaces investigated. Numerous physical contacts of the surface during desorption did not significantly affect the desorption process.

INTRODUCTION

The adsorption of tritium gas on various surfaces and the consequent health hazard to personnel who might contact these surfaces has been of concern to this laboratory for several years. It is well known that when objects or materials are exposed to an environment containing tritium, their surfaces become contaminated.

In order to determine how much activity is adsorbed on and desorbed from these materials, conventional sorption studies are required. In addition to the determination of sorption phenomena, studies of the amount of activity removed by direct contact with the surface are necessary to determine the hazard involved.

The principal impediment to performing these measurements has been the lack of proper sampling instrumentation. A swabbing technique was developed some time ago.[1] That technique did not provide the reproducibility needed to intercompare samples taken consecutively over the same area of contaminated surface. Furthermore, an intercomparison of the surface contact desorption process between surfaces of different materials and coatings was difficult with the swabbing technique.

This report describes and evaluates an apparatus which can be used to measure the amount of activity that would be removed from a contaminated surface by directly contacting the surface. These measurements make possible studies of surface contact desorption that would provide information for assessing situations where personnel health hazards may exist.

APPARATUS AND PROCEDURE

The apparatus consists of three basic parts; (1) the support frame, (2) the carriage and (3) the sampling roller. The complete apparatus is shown in operation in Fig. 1. The principle of the apparatus will be presented by describing a measurement on a typical sample.

The surface to be sampled (in the present case a plate 3 X 10 X 1/4 in.) is exposed for a specific time to a known concentration of tritium in an exposure system shown in Fig. 2. The contaminated plate is then removed and placed on the sample plate holder on the base of the support frame (Fig. 3).

The sample plate holder is designed so that only four points of the contaminated surface are contacted. This feature permits sampling of both sides of the plate without cross-contamination. The support pins are spring-loaded to keep the plate surface at a fixed level with respect to the sampling roller.

A strip of No. 42 Whatman filter paper is wrapped tightly around the glass roller and is fastened to the roller by a glass insert that holds the ends of the paper in a slot in the roller (Fig. 4). After assembly the roller is dipped in ethylene glycol and blotted to dampness. A supply of assembled rollers can be stored under ethylene glycol for later use (Fig. 5). The pertinent glass roller data is summarized in Table 1.

TABLE 1

Pyrex Glass Roller Data

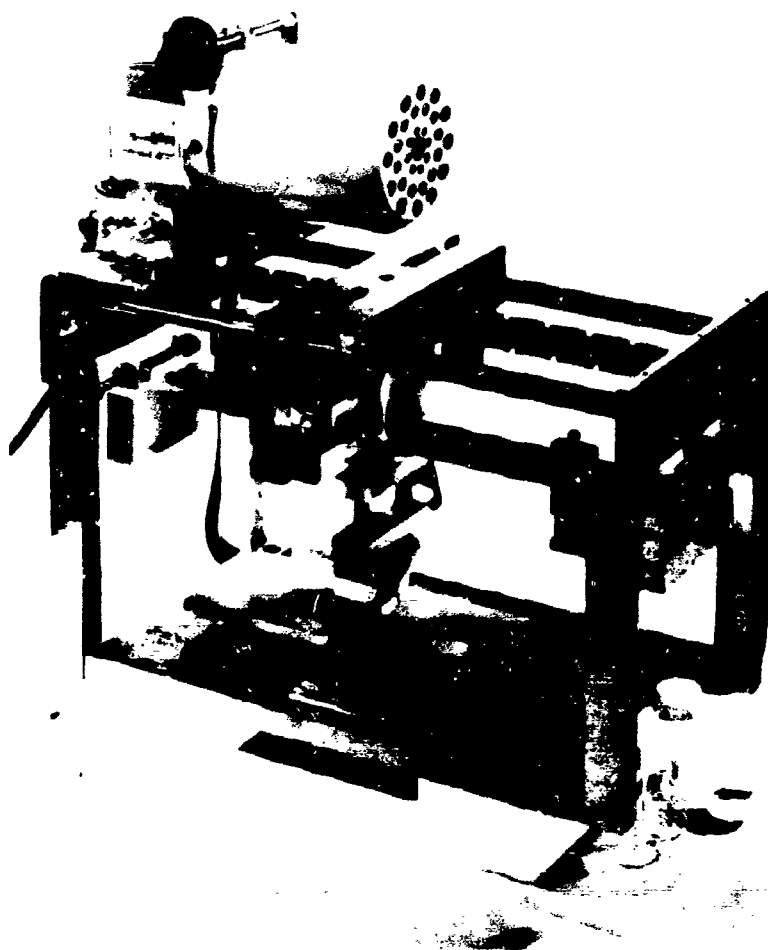|	
Roller Diameter	1.00 cm
Roller Body Length	2.00 cm
Pyrex Glass Insert Thickness	0.13 cm
Effective Sample Length per Revolution	3.01 cm (3.14–0.13)
Effective Sample Area per Revolution	6.02 cm^2

2

Fig. 1 Apparatus in Operation. The sampling roller is being rolled across a strip of contaminated painted steel plate.

VACUUM

(VACUUM STOPCOCKS ARE
TEFLON AND VITON)

TRITIUM
SUPPLY

GAS PIPETTE

0.1

AIR

0.5

MERCURY
RESERVOIR

1.0

SAMPLE PLATE

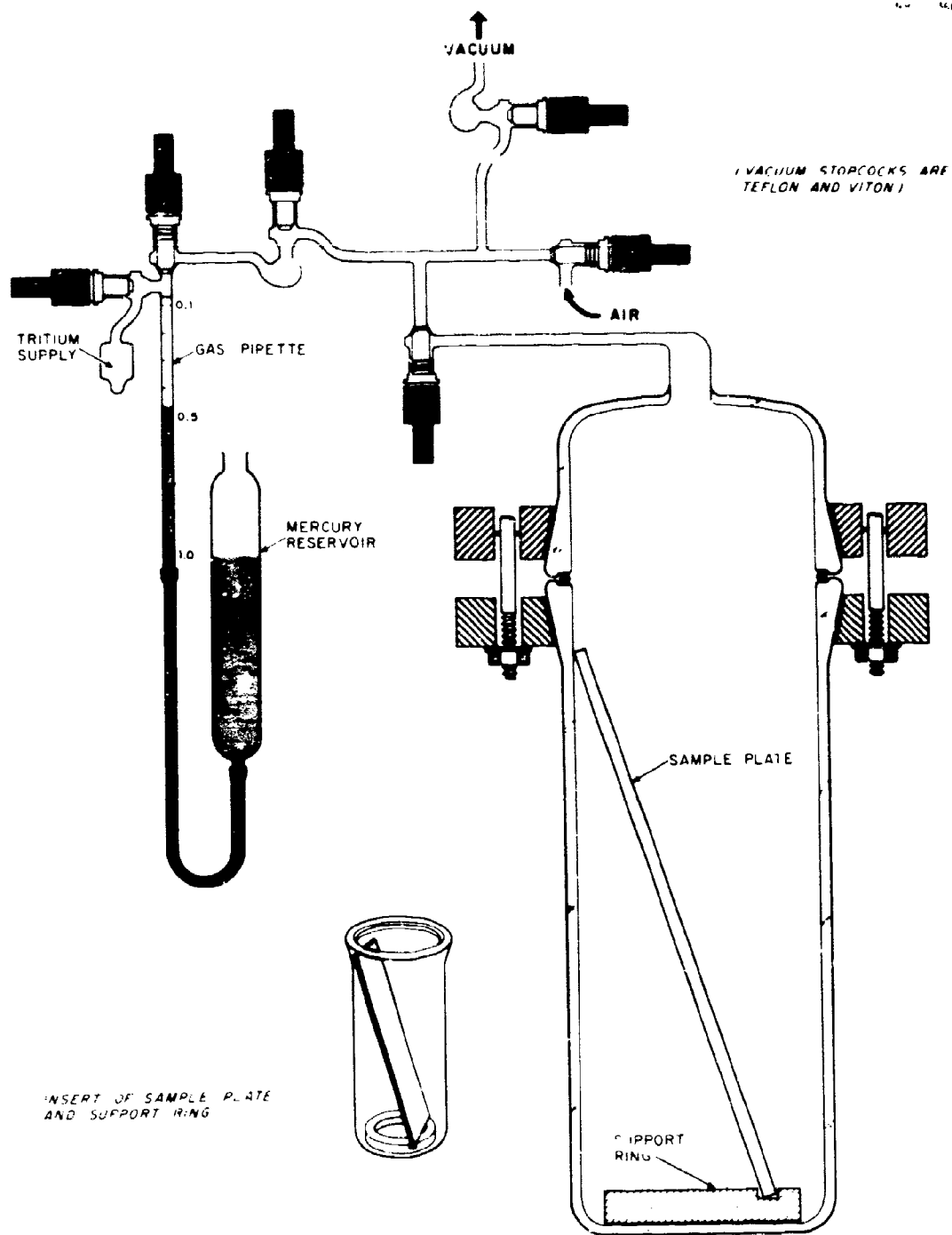INSERT OF SAMPLE PLATE
AND SUPPORT RING
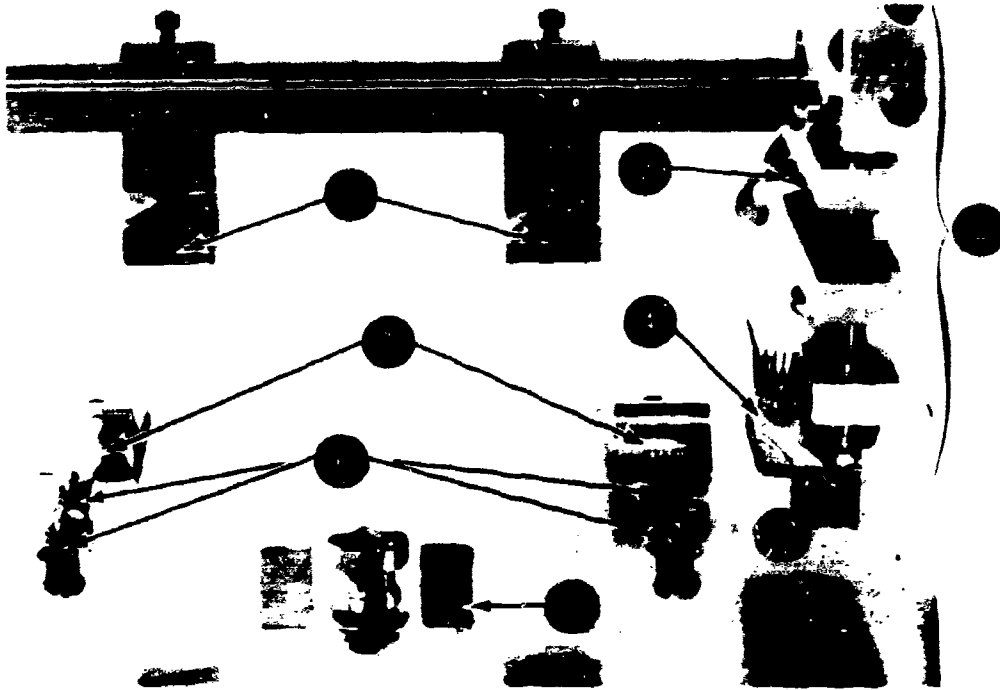
SUPPORT
RING

Fig. 2 Exposure System

Fig. 3 Lower Portion of the Apparatus

1. Cam actuators.
2. Cam.
3. Roller carriage.
4. Roller retainer.
5. Glass roller.
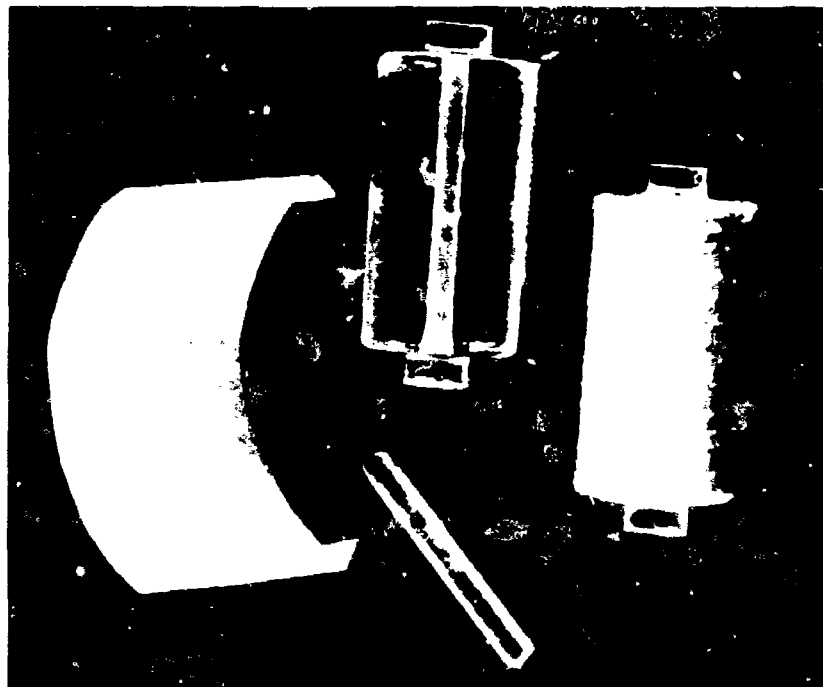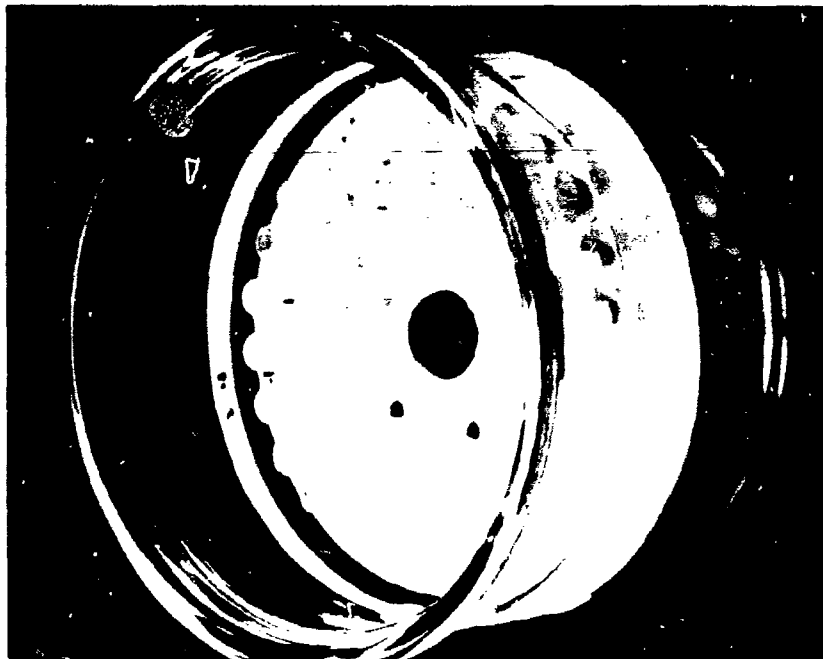6. Spring-loaded plate support pins.
7. Plate clamp.

Fig. 5 Roller Storage Container. The rollers are positioned end up with one axel supported in a hole in the porcelain desiccator. The solvent is ethylene glycoi.

Fig. 4 Sampling Roller Components. The glass insert holds the filter paper on the roller.

Preparatory to sampling, a roller is placed in position on the bottom of the carriage (Fig. 6) and secured by means of the roller retainer. The carriage and its lateral support rods are rigidly suspended perpendicularly from two stainless steel guide rods and a two-way drive screw (Fig. 1). The lateral support rods (Fig. 6) allow for lateral positioning of the carriage such that three adjacent strips can be sampled (consecutively) on one side of the contaminated plate. Two indents on the lateral support rod serve to reproducibly position the sampling roller over adjacent strip positions.

The guide rods and the two-way drive screw are mounted on the aluminum support frame. The two-way drive screw is driven by a small variable speed motor which is started by a manual microswitch. The "Speedmate" motor used was able to drive a carriage over the sample plate at speeds ranging from 0.50 to 10 cm/sec. Two other microswitches wired to the motor are mounted at opposite ends of the aluminum support frame. They are normally closed during movement of the carriage. One or the other is opened, thereby shutting off the motor when depressed by the carriage as it comes against the support frame at the end of a run. The manual switch is again needed to restart the drive motor.

The glass sample roller is lowered to the surface of the contaminated sample plate by a unique cam mechanism shown in Fig. 7. This mechanism also lifts the sample roller off the plate surface after the run. The positions where the roller is lowered onto and lifted off the plate can be selected by positioning the cam actuators along a bar that extends the horizontal length of the support frame (Figs. 3 and 11).

When the sample roller has contacted the plate surface and reached the end of the frame, at the end of a run, the roller retainer is opened manually (Figs. 3 and 8) and the roller drops directly into a counting vial containing scintillant solution. The time elapsed between completion of surface sampling and immersion of the contaminated roller in the scintillant solution is less than 5 sec. The counting sample is shown in Fig. 9. A dioxane scintillant solution[2] is used for tritium determination. The formula for this solution is presented in Table 2.

The weight that the glass roller exerts on the sample surface is determined by a spring-loaded mechanism on the roller carriage. The spring tension can be varied over a limited range by means of a thumb screw (Fig. 6). The overall range can be changed by replacing the springs with others of different tension. A contact weight range of 1.50 to 5.0 lb was available using the apparatus described in this work. A weight of 5 lb was arbitrarily chosen for the contact runs presented. The surface contact rate used was 2.2 cm^2/sec and the contaminated area contacted per sample was 14.4 cm^2.
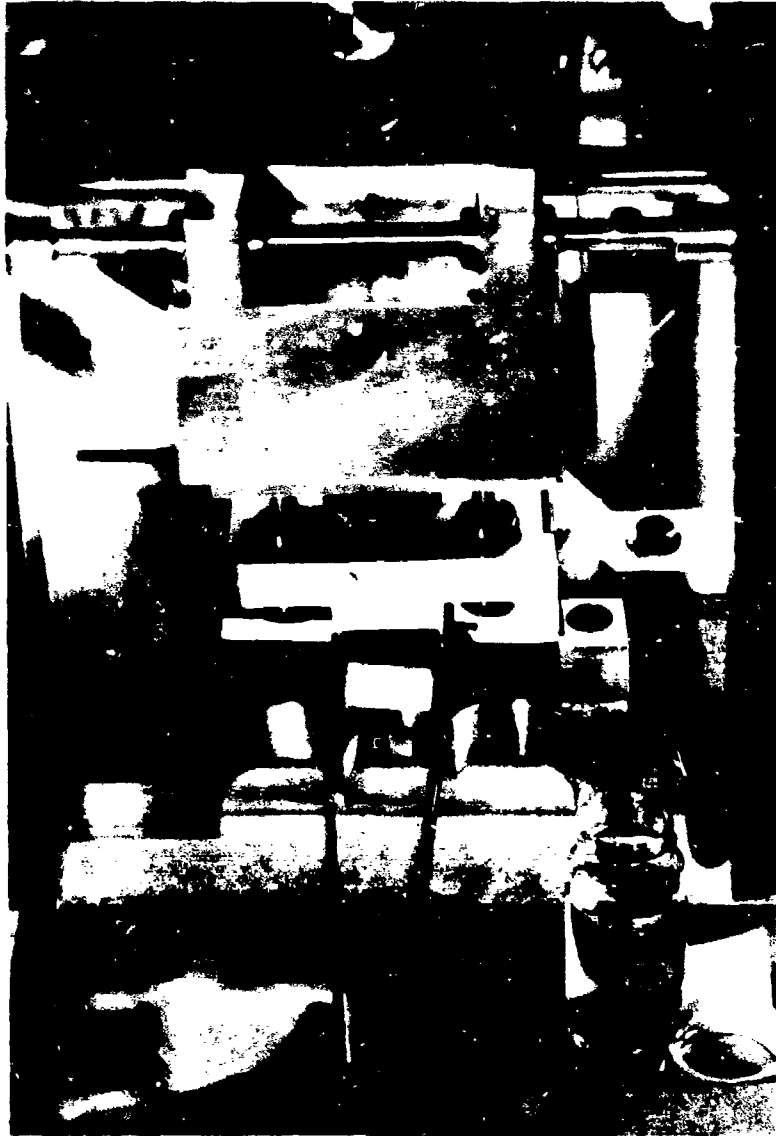
Fig. 6 Loading the Glass Roller. The left hand is used to hold open
the roller retainer (out of view behind the rider; see Fig. 3). The
thumb screw on the rider above the roller bracket is used to adjust the
weight of the roller against the plate.

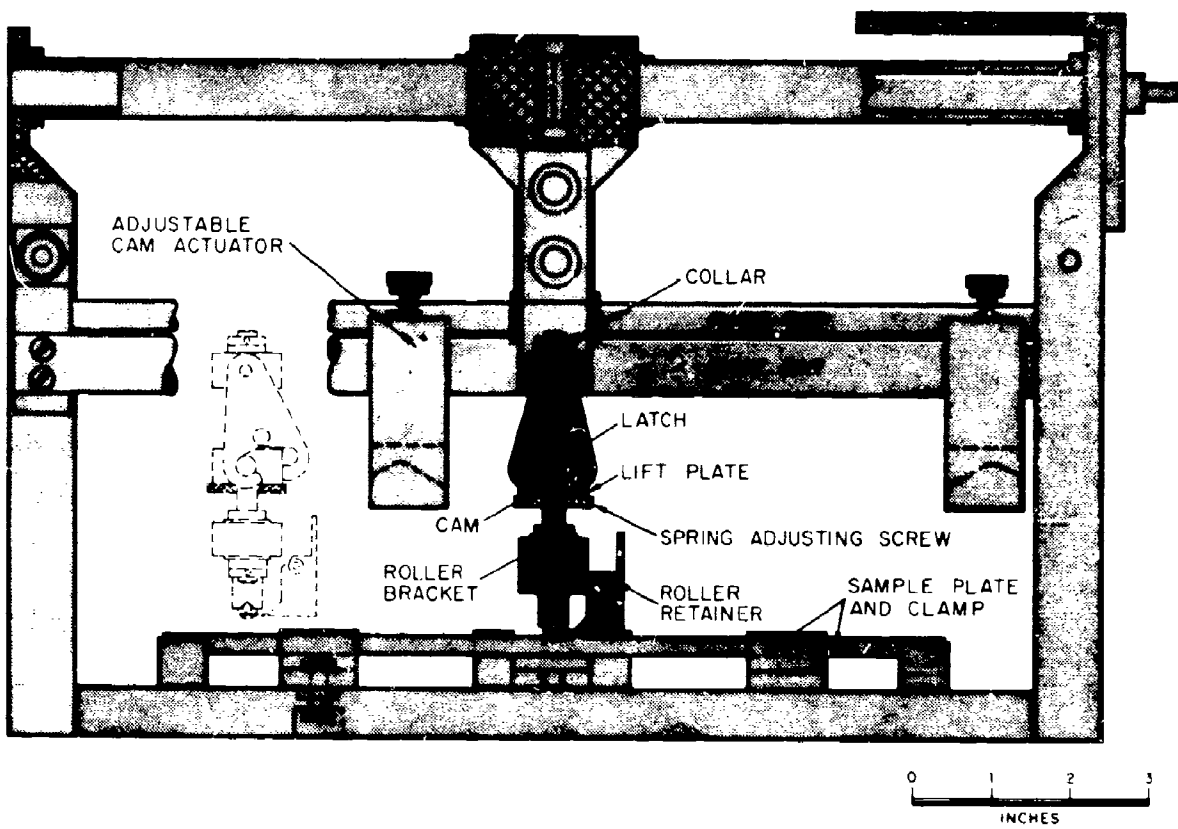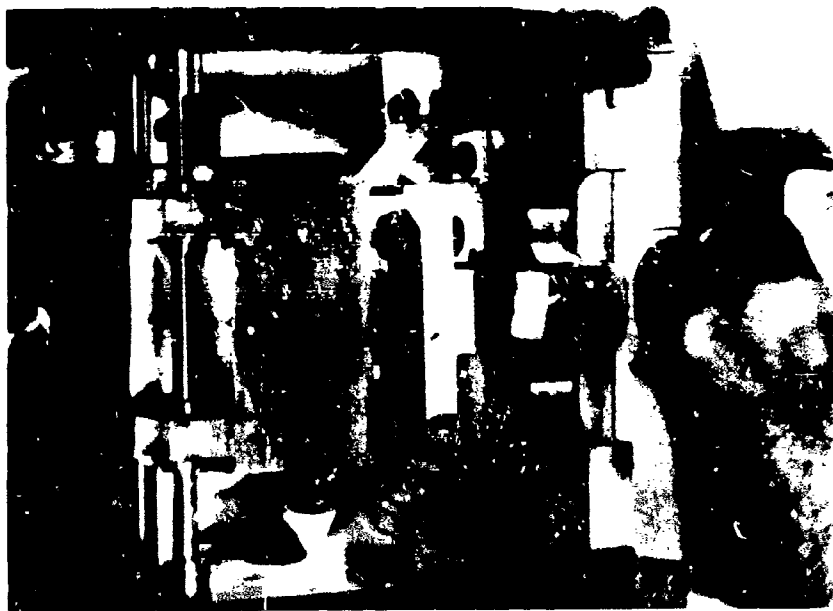Fig. 7 Cam Mechanism

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Fig. 8 Release of Roller. The roller retainer (out of view behind the rider; see Fig. 3) has been depressed to release the roller into the sample bottle containing scintillant solution.
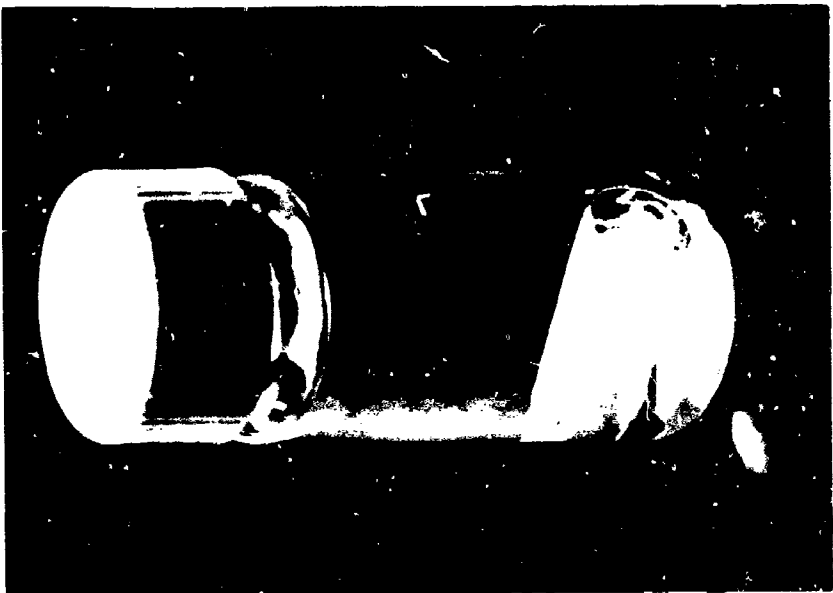
Fig. 9 Scintillation Sample. The roller contaminated with radioactivity rests horizontally on the bottom of the scintillant bottle and provides a reproducible counting geometry.

TABLE 2

Formula for Scintillator

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60 g - Naphthalene (Recrystallized)

4 g - 2,5-Diphenyloxazole,[PPO],(Scintillation Grade)

*0.2 g - 1,4-bis-2-(4-Methyl-5-Phenyloxazolyl)-Benzene, [Dimethyl POPOP]
      (Scintillation Grade)

100-ml - Methanol, Abs. Anal. Reagent

20 ml - Ethylene Glycol, Anal. Reagent

Dissolve the above materials in pure p-Dioxane and dilute to 1 liter
volume with more p-Dioxane.

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*Dimethyl POPOP was substituted for the POPOP used in Bray's original
formula.

The dimensions of the apparatus are such that with the motor removed
the apparatus will pass through the door (7-1/2 × 9-1/2 in.) of a con-
ventional glove box, and that after assembly the apparatus can be easily
operated therein.

In order to correlate surface counting with the amount of activity
removed from the surface by contact, a slide chamber (Fig. 10) and a
special planchet plate (Fig. 11) are used. The contaminated planchet is
first counted in the chamber, then placed in a cut-out on the adapter
plate and the surface is contacted under fixed conditions. The amount
of activity removed is determined by scintillation counting as previously
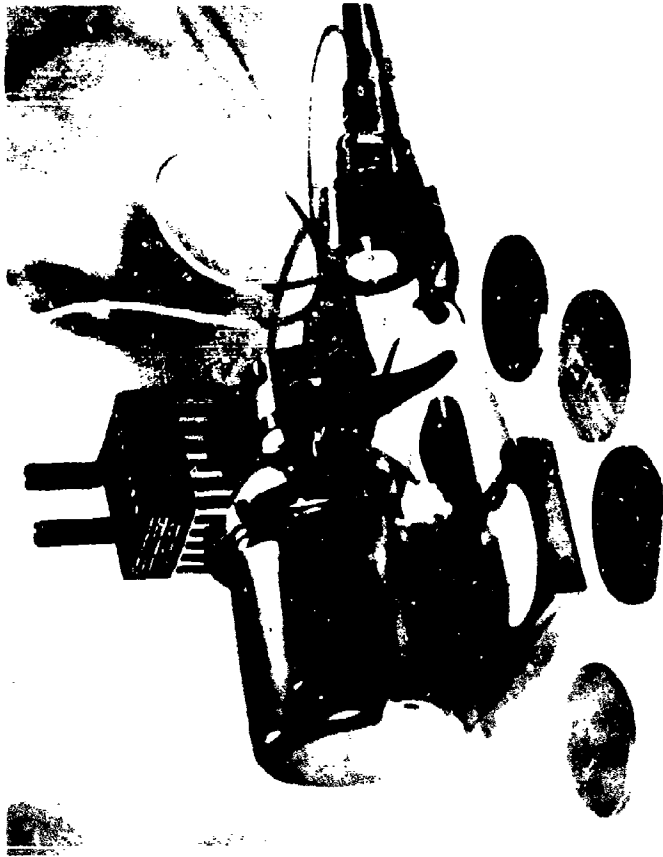described, and the planchet is recounted in the chamber.

Fig. 10 Slide Chamber. The chamber is
made portable for use near the exposure
vessel by using a shielded connector
which is adapted for a coaxial cable
from a micro-micro-ammeter.
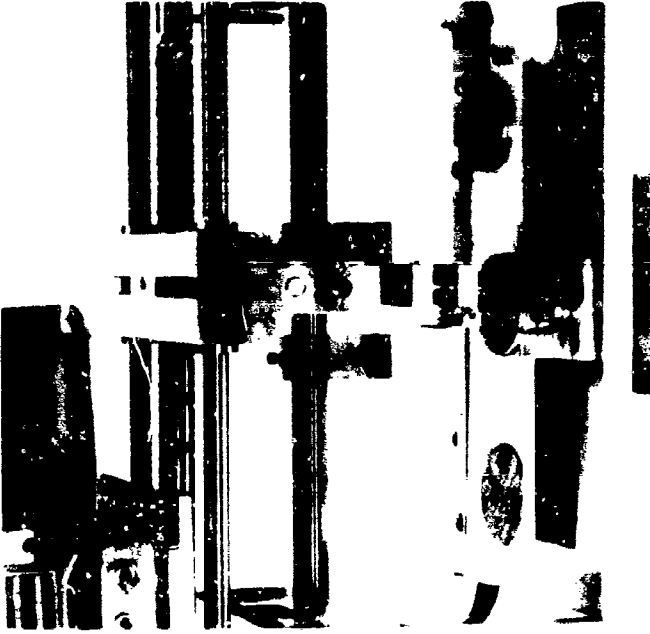

Fig. 11 Planchet Sampling. The slide-
chamber-planchet adapter plate instal-
led in the sample plate position. Note
how the cams are positioned to sample
only the center planchet. There are
cut-outs on the plate to position the
planchets.

EVALUATION OF APPARATUS

Determination of Sampling Precision

The sampling precision of the apparatus was evaluated by placing known amounts of non-volatile activity, Pm^{143}, on a specific area of surface and then removing a portion of the activity by contact with the sample roller.

The material used for the precision test was a plate of optical quartz. The surface was degreased with alcohol, washed with distilled water, and dried in an oven at $200^{\circ}C$. The plate was removed and brought to room temperature. 2×10^5 d/m of Pm^{143} (in dilute HCl) were carefully pipetted onto each of five strips marked out on the surface of the plate. Pipetting was so done that the small drops of activity were distributed uniformly within the marked strips. The contaminated surface was dried carefully in air. Each strip, containing a known amount of contaminant, was then sampled with a fresh sample roller. A precision of $\pm 2 \%$ was obtained for the five measurements.

Poorer precision was obtained in cases of sampling relatively coarse-textured nonuniform surfaces contaminated with tritium activity. Runs made with a washed carbon steel plate exposed to a tritium-containing atmosphere gave a precision of $\pm 25 \%$. Runs made on the reverse side of the same plate using a swabbing technique gave a precision of $\pm 65 \%$. The poorer precision obtained with the swabbing technique is mainly due to the swabs' sampling unequal areas.

The large difference in precision between the quartz surface and the metal surface is attributed primarily to the observed nonuniformity of the latter surface. The possible variation in tritium volatility on different areas of a given contaminated metal was not independently determined.

Effect of Ethylene Glycol Evaporation from Sample Roller

In order to determine the ethylene glycol loss on the roller and to evaluate its effect on the amount of tritium removed by contact, as well as the effect of water absorbed by the ethylene glycol on the counting sensitivity of the scintillant, the following measurements were made. The weight loss in air of ethylene glycol from a dampened roller was determined as a function of time and found to be less than 2 % for open air evaporation during the evaporation period from 60 to 300 minutes

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after dampening (Fig. 12). This small loss of ethylene glycol did not measurably affect the surface contact sampling results. Ethylene glycol was used as a component of the scintillant solution (Table 2).

Initially the ethylene glycol on the roller will absorb some water if it is not already at equilibrium with the water vapor in the air. The magnitude of this absorption is shown by the initial portion of the curve in Fig. 12. No measurable quenching of the scintillant due to the small amount of water absorbed in the glycol was observed.

Performance Test Runs

To determine whether the apparatus could be used to measure the effect of a dirt-oil coating on tritium adsorption, surface contact measurements were made on both a washed and an unwashed surface. The structural steel plates obtained for use as samples had a coating of dirt and oil on each side. The results are shown in Table 3. It appears that there are several orders of magnitude more of tritium activity removed from the dirty surface than from a clean surface. This is probably because much more tritium is initially adsorbed by the dirtier surface.

Measurements were also made on a third (unwashed) sample which had noticeably more dirt on one side than the other. It was determined for this sample that significantly more tritium was adsorbed on the dirtier side. The effect of different exposure times on the results was not determined. However, it has been established that there is no significant change in the total amount of tritium adsorbed on a washed structural steel surface when exposure times were varied from 15 min to 4 hr.*

Other measurements performed with the apparatus consisted of sampling different surfaces as a function of time after exposure to tritium. Data for these surfaces and their coatings are listed in Table 4. The general procedure followed was to multiply sample one area of the surface with fresh rollers at various times after exposure, while sampling other areas of the same surface only once at intervals spread over the total sampling period (usually 3 hr). The results of these "contact desorption" measurements for pyrex glass, structural steel, carbon steel, and various surface coatings are summarized in Figs. 13-19.

The results for the pyrex glass surface show that three orders of magnitude less of tritium activity is removed by contact than from the other surfaces. It was not determined whether less tritium activity

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*W. R. Balkwell, D. A. Kubose, "The Determination of the Total Tritium Activity Adsorbed on the Surface of Various Metals as a Function of Tritium Exposure Concentration and Exposure Time," U. S. Naval Radiological Defense Laboratory, USNRDL-TR, to be published.
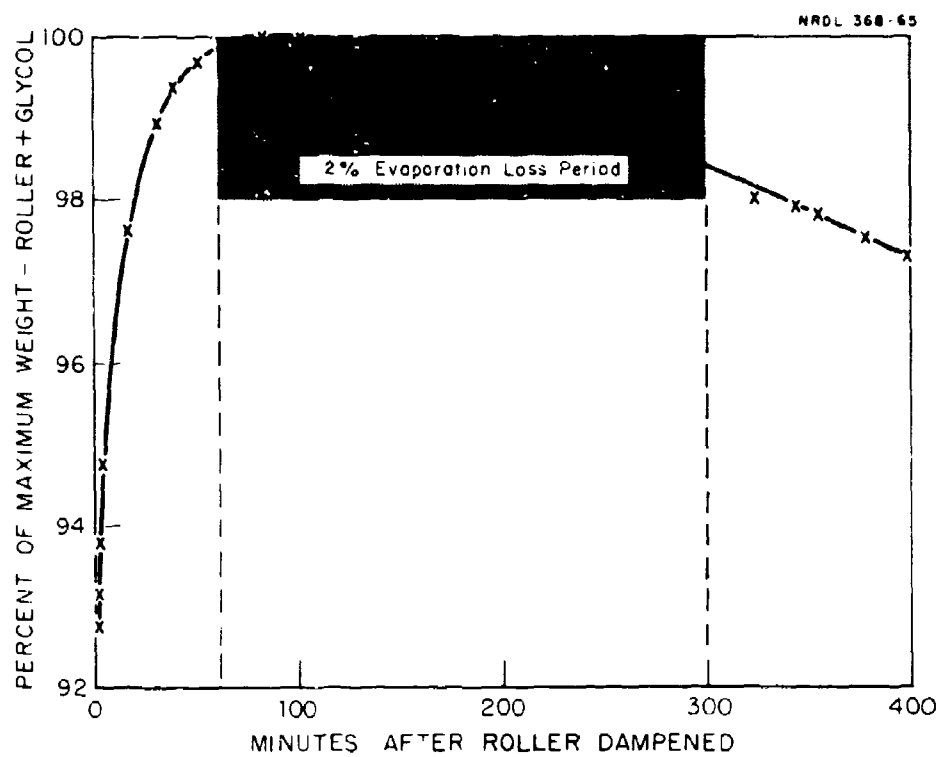
Fig. 12 Evaporation of Ethylene Glycol From Sample Roller

Table 3

SAMPLE DATA

| Material | Plotted Point | Washing Procedure | Surface Coating | | | Tritium Pumped out Before Sample Removed From Exposure Vessel | Tritium Exposure Concentration mcT$_2$/cc | Exposure Time Hours |
			First Coat	Second Coat	Third Coat			
Pyrex glass	✚ ✖	Alcohol-water	-	-	-	Yes	0.046	96
Structural steel	■ □	Toluene Methanol Hot water	-	-	-	No	0.076	1
Carbon steel	◆ ◇	Toluene Methanol Hot water	-	-	-	No	0.061	1
Structural steel	● ○	Toluene	Zinc Chromate #84	Zinc Chromate #84	-	Yes	0.054	24
Structural steel	▶ ▷	Toluene	Devran #202	Devran #203	Devran #203	Yes	0.031	48
Structural steel	● ○	Toluene	Zinc Chromate #84	Red Lead #116	-	Yes	0.044	24
Structural steel	◀ △	Toluene	Wash Primer #117	Red Lead #116	-	Yes	0.047	24
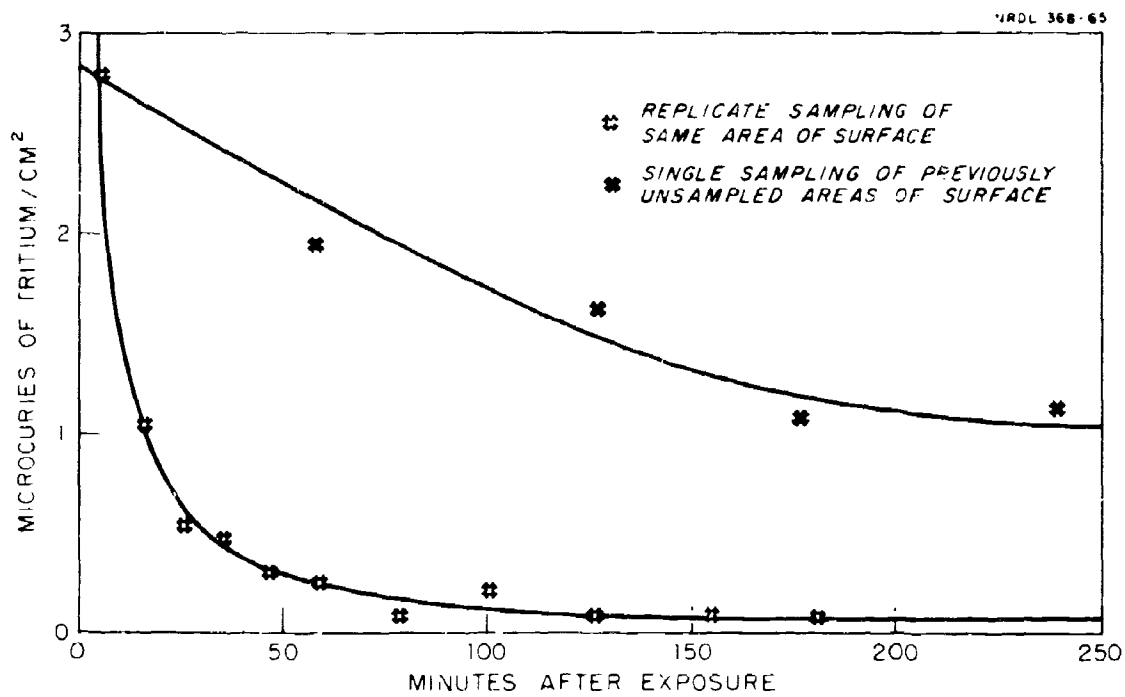
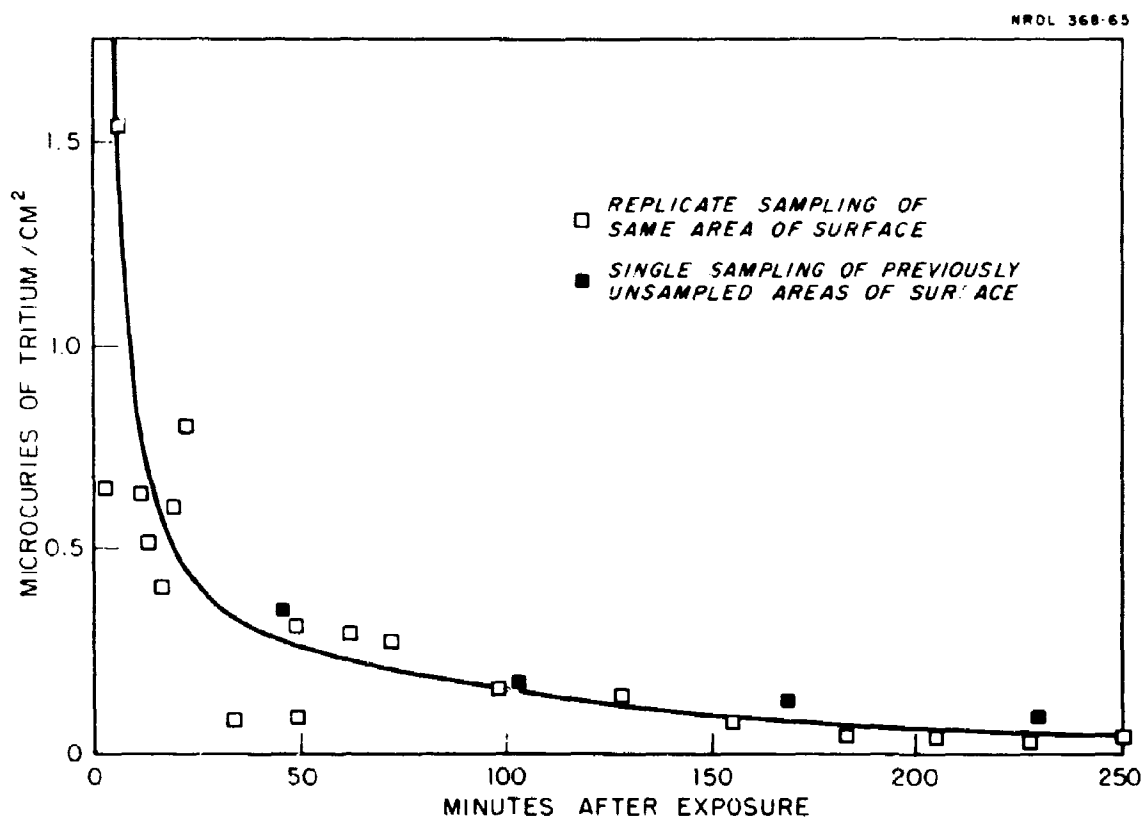Fig. 13 Contact Desorption of Tritium From Glass

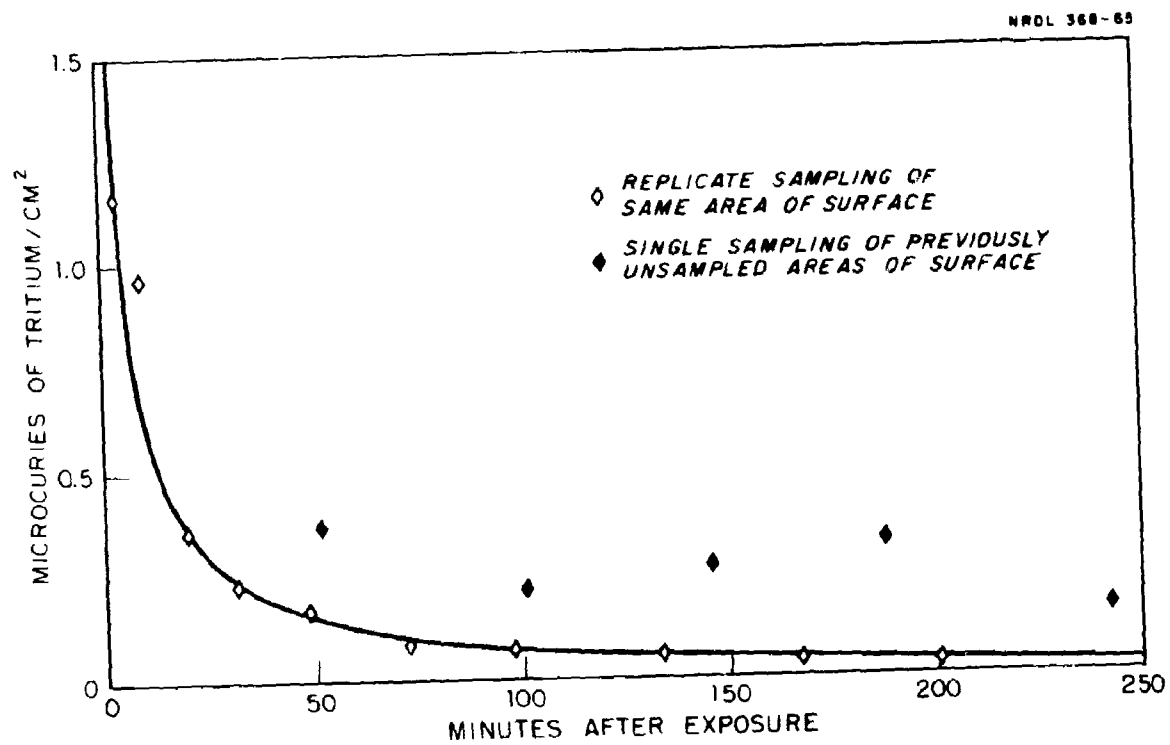Fig. 14 Contact Desorption of Tritium From Structural Steel

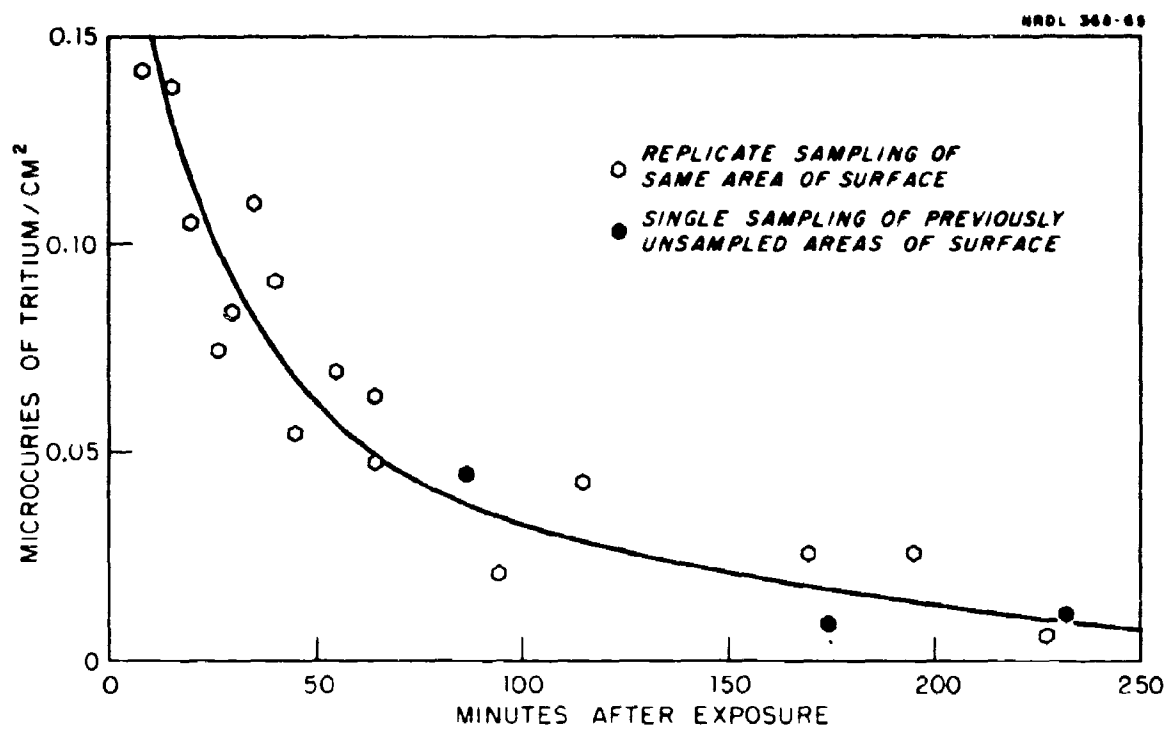Fig. 15 Contact Desorption of Tritium From Carbon Steel

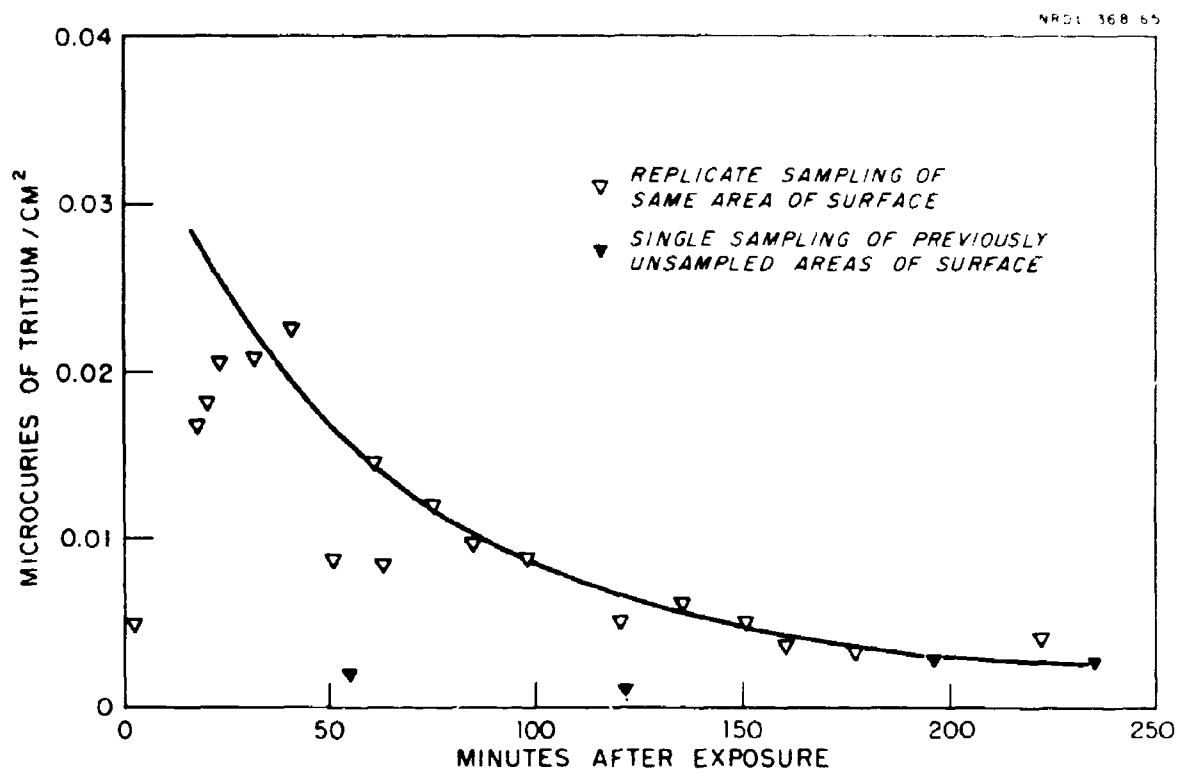Fig. 16 Contact Desorption of Tritium From Painted Structural Steel

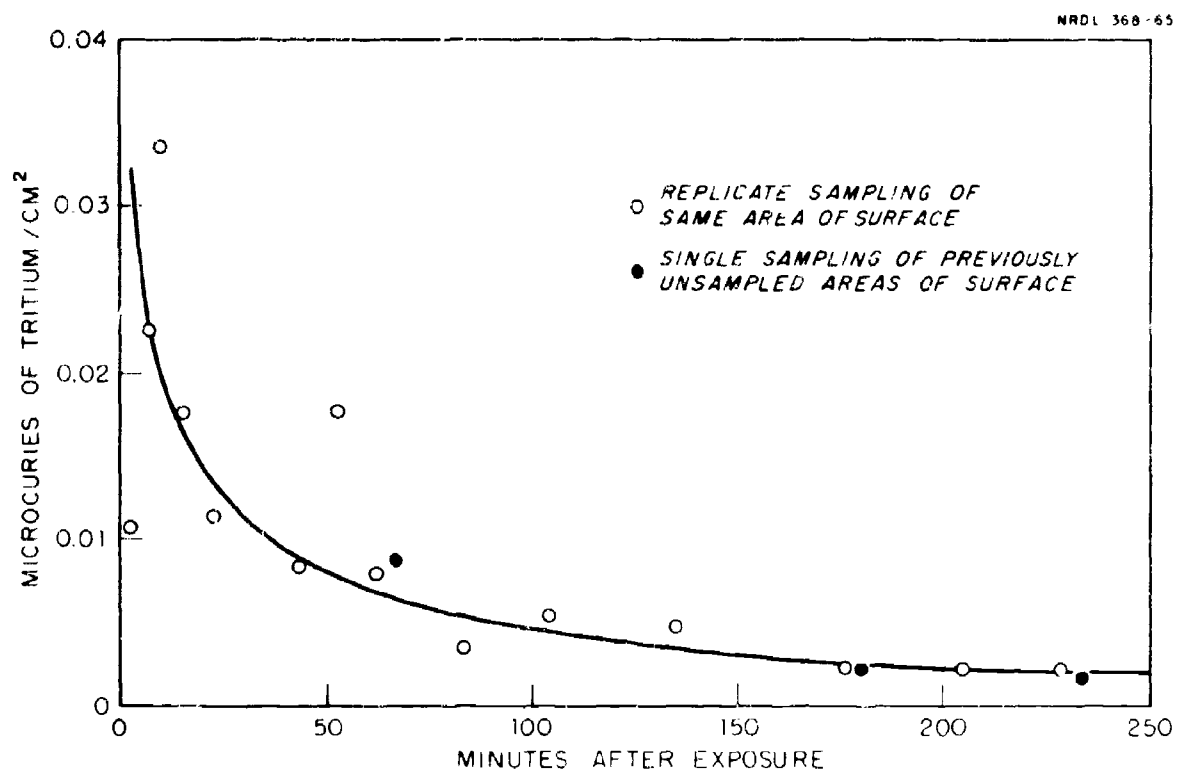Fig. 17   Contact Desorption of Tritium From Painted Structural Steel

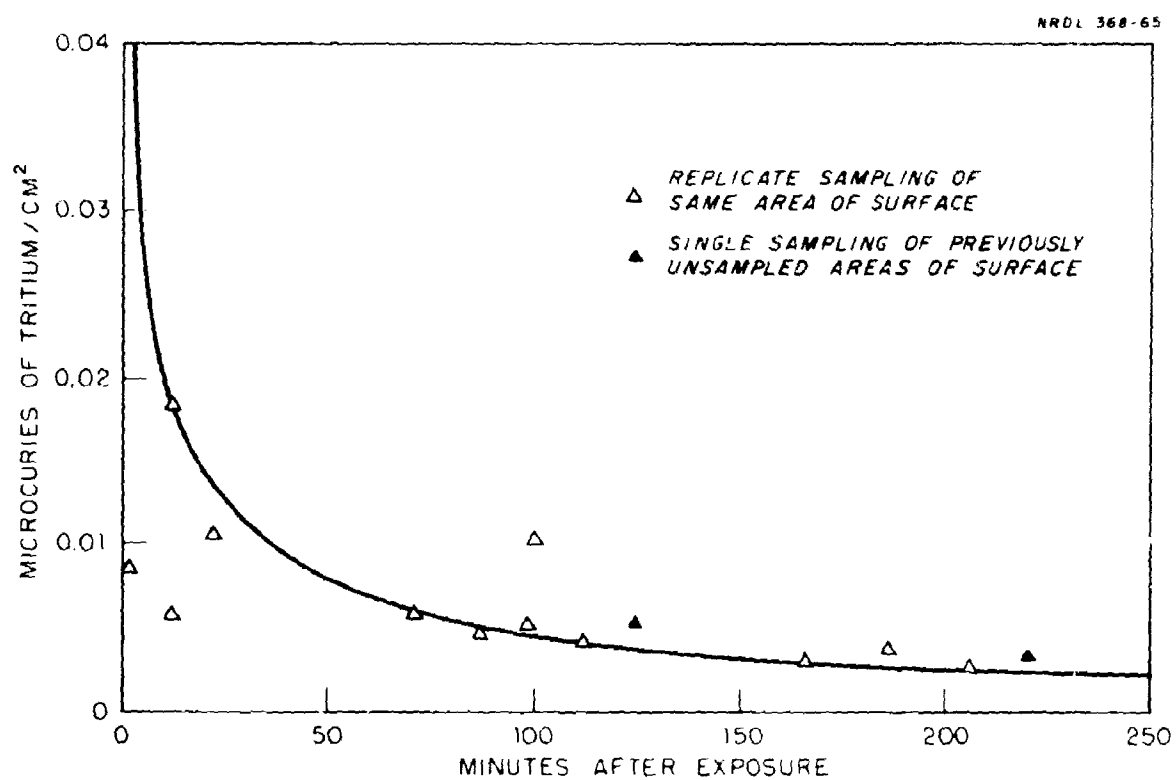Fig. 18 Contact Desorption of Tritium From Painted Structural Steel

Fig. 19 Contact Desorption of Tritium From Painted Structural Steel

TABLE 3

Tritium Contact Removal From Washed and Unwashed Surfaces of Structural Steel

Sample	Toluene and Hot Water Wash Before Exposure	Original* Exposure Concentration (mc T_2/cm^3)	Exposure Time (days)	Amount of Tritium Removed for a Single Contact with Surface ($\mu c/cm^2$)
1	yes	0.028	3	$9 \ \times \ 10^{-6}$
2	no	0.028	1	1.2×10^{-2}

*Each contaminated sample flushed with several atmospheres of clean air before sampling surface.

is initially adsorbed on pyrex. The relative removal of tritium activity by "contact desorption" compared to that by desorption alone is greater for pyrex than for the metal and painted surfaces.

For the metal and painted surfaces the single contact and multiple desorption processes were essentially identical, i.e., multiple contacting of the surface did not significantly alter the inherent desorption process. The amount of tritium activity removed by contact from the painted surfaces is about an order of magnitude less than that removed from the bare metal surface. Again, it was not established whether this was due to less initial adsorption of the activity on the painted surfaces relative to the bare metal surface.

CONCLUSION

The results presented in this report are representative of the kind of measurements that can be performed with the apparatus described. The amount of tritium removed from a surface by direct contact can be determined for single or multiple contacts with the contaminated surface. Surface contact desorption measurements can be made to determine the time required to attain surface contact desorption equilibriur. Evaluation

of the efficiency of various decontamination procedures from the standpoint of removal of surface contamination can also be studied. The effectiveness of various solvents for adsorption of surface activity may also be analyzed by varying the solvent on the roller. No doubt, there may be other unforeseen applications of this apparatus, particularly in the field of health physics.

REFERENCES

1. A. E. Symonds, Jr., "Removal of Tritium Contamination From Surfaces of Metals," AEC Research and Development Report, DP-367, 1959.

2. G. A. Bray, "A Simple Efficient Liquid Scintillator for Counting Aqueous Solutions in a Liquid Scintillation Counter," Anal. Biochem. 1:279, 1960.